07/02/102

ON - LINE TEXT VALIDATION

PAGE 1

17:37:03 ERROR REPORT

PATENT NO: 56055692.001 GROUP: T1 ISSUE DATE: 06/10/02 PAGE NO: 2 LINE NO: 12 ERROR => NO SECTION COMMAND FOUND AT START OF TEXT
PAGE NO: 2 LINE NO: 24 WARNING => MISSING LIGHT COMMAND (+1) FOLLOWING (+b) AFTER 50 !! PAGE NO: 11 LINE NO: 227 WARNING => INFER/SUPR COMMANDS MUST RETURN TO LIGHT AFTER 10 PAGE NO: 11 LINE NO: 227 WARNING => INFER/SUPR COMMANDS MUST RETURN TO

LIGHT AFTER 10

DACS-ERROR REPORT

PATENT	Г #: =====	5605! ====	5692	2.001	- -	IS	SUE	DATE	: 06/	10/02			GROUP:	T1 ======

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- 0001 +pg,1
- 0002 +sa
- 0003 An ethylene/+60 -olefin polymer blend is described comprising first <<<<>>>>and second ethylene/+60 -olefin
- 0004 polymer components in which the ethylene content of the first <<<<>>>>component is at least 10
- 0005 weight percent different than the ethylene content of the second <<<<>>>>component. These blends
- 0006 exhibit an improved combination of low temperature, pellet flow, <<<<>>>>compression set, melt strength
- 0007 and/or shape retention properties as compared to either component, or <<<<>>>>an ethylene/+60 -olefin
- 0008 polymer blend of similar composition but in which the ethylene <<<<>>>>content of each component is
- 0009 substantially the same.
- 0010 +ea

- 0011 + pg, 2
- 70012 to +p This application claims priority from Provisional application Ser. <<<>>>>No. +b 60/203,298, +1
- 0013 filed May. +b 11, 2001.
- 0014 +su +cl FIELD OF THE INVENTION
- 0015 +p This invention relates to ethylene/+60 -olefin polymer blends. In <<<<>>>>one aspect, this invention
- 0016 relates to polymer blends comprising two or more ethylene/+60 -olefin <<<< >>>>components while in
- 0017 another aspect, this invention relates to blends in which one or more <<<<>>>>of the components
- 0018 comprises an ethylene/+60 -olefin/polyene polymer. In yet another <<<<>>>>aspect, this invention relates to
- 0019 polymer blends of ethylene/+60 -olefin components in which the <<<< >>>ethylene content of one
- 0020 component differs from the ethylene content of at least one other <<<<>>>>component by at least about +b 10 +l
- 0021 weight percent.
- 0022 +cl BACKGROUND OF THE INVENTION
- 0023 +p Ethylene/+60 -olefin polymer blends are well known in the art. The <<<
>>>blends taught in U.S. Pato Nos.
 - 0024 +b 4,438,238; 4,722,971; 4,874,820; 4,902,738; 4,937,299; 4,939,217; <<<<>>>>5,013,801; 5,236,998;
 - 0025 5,292,845; 5,382,631; 5,494,965; 5,539,076; 5,691,413; 5,728,766; 4,<<<<>>>>429,079; 4,530,914;
 - 0026 5,605,969; 5,338,589; 5,260,384; 5,478,890; 5,438,100; 5,476,903; 5,<<<<>>>>703,180; 5,464,905;
 - 0027 5,744,551; 5,747,620 +1 and +b 5,798,427 +1 are representative, and <<<<>>>>each of these patents are

- 0028 incorporated herein by reference.
- 0029 +p Blends are useful because they provide properties not available <<<< >>>>from the individual
- 0030 components from which the blend is made. For example, an ethylene/+60 <<<<>>>>-olefin polymer with a
- 0031 relatively narrow molecular weight distribution (MWD), e.g., +b 2 +l <<<<>>>>or less, will usually produce a
- 0032 film with good transparency but it will usually process less <<<<>>>>efficiently than an ethylene/+60 -olefin

- 0033 + pg, 3
- 0034 polymer alike in all aspects except with a MWD of +b 3 +l or more. <<<>>>>However, an ethylene/+60 -olefin
- 0035 polymer with a MWD of +b 3 +l or more usually produces a film that is <<<< >>>>less transparent than a like
- 0036 ethylene/+60 -olefin polymer with a MWD of +b 2 +l or less. Blending <<<< >>>>the two polymers will usually
- 0037 produce a composition that will produce a film with both desirable <<<< >>>>transparency and
- 0038 processability. Moreover, depending upon the particular ethylene/+60 -<<< >>>olefin polymers, the relative
- 0039 proportions of each, the manner in which the polymers are made and/or <<<< >>>>blended, the properties
- 0040 of interest and a host of other variables, one or more properties of <<<<>>>>the blend may be more than a
- 0041 simple average of its component parts.
- 0042 +p While ethylene/+60 -olefin polymer blends can be prepared by any <<<<>>>>one of a number of
- 0043 different processes, generally these processes fall into one of two <<<<>>>>categories, i.e., post-reactor
- 0044 blending and in-reactor blending. Illustrative of the former are melt <<<<>>>>extruders into which two or
- 0045 more solid ethylene/+60 -olefin polymers are fed and physically mixed <<<>>>>into a substantially
- 0046 homogeneous composition, and multiple solution, slurry or gas-phase <<<<>>>>reactors arranged in a
- 0047 parallel array the output from each blended with one another to form <<<<>>>>a substantially
- 0048 homogeneous composition which is ultimately recovered in solid form. <<<<

- >>>>Illustrative of the latter
- 0049 are multiple reactors connected in series, and single reactors <<<< >>>>charged with two or more catalysts.
- 0050 While each general process category has its own advantages and <<<<>>>>disadvantages, in-reactor
- 0051 blending is a favored technique for making blends in which component <<<<>>>>compatibility, i.e., the
- 0052 ability to make a substantially homogeneous blend from the components,<<<<>>>> is a factor. Generally,
- 0053 forming a substantially homogeneous blend from ethylene/+60 -olefin <<<<>>>>polymer components that are
- 0054 less than fully compatible is easier and more successful and cost <<<<>>>>effective using an in-reactor
- 0055 technique than a post-reactor technique, particularly melt extrusion.

- 0056 + pg, 4
- 0057 +p Ethylene/+60 -olefin polymers and blends of these materials are <<<<>>>>commercially important
- 0058 because they exhibit and/or impart desirable properties to various <<<<>>>>products, e.g., films and
- 0059 molded and extruded articles. Properties of frequent interest are low <<<<>>>>temperature impact
- 0060 strength, compression set, melt strength, shape retention, pellet <<<< >>>>flow, mechanical strengths and
- 0061 modulus. Depending upon the end use, often one or more of these <<<< >>>>properties will be more
- 0062 important than the others. Enhancement of these more important <<<<>>>>properties often requires the
- 0063 use of a blend of ethylene/+60 -olefin polymers. The industry <<<<>>>>interest, of course, is in blends in
- 0064 which the properties of primary importance are enhanced without <<<< >>>>significant diminution of the
- 0065 other properties.
- 0066 +cl SUMMARY OF THE INVENTION
- 0067 +p According to this invention, ethylene/+60 -olefin polymer blends <<<<>>>>with improved low
- 0068 temperature, pellet flow, compression set, melt strength and/or shape <<<<>>>>retention properties are
- 0069 prepared by blending a first ethylene/+60 -olefin polymer component <<<<>>>>with a second ethylene/+60 -olefin
- 0070 polymer component, with the proviso that the ethylene content of the <<<< >>>>first and second
- 0071 ethylene/+60 -olefin polymer components differ from one another by at <<<>>>>least about +b 10 +l weight

- 0072 percent. The blends can be made by either post-reactor or in-reactor <<<<>>>>blending, and the weight
- 0073 ratio of first component to second component can vary widely, <<<<>>>>typically from between +b <math>80:20 +l to
- 0074 +b 20:80. +l One hallmark of this invention is that the enhanced <<<<>>>>properties of the blend are achieved
- 0075 without significant diminution of other desirable properties of the <<<<>>>>blend components.

- 0076 + pg, 5
- 0077 +dr +cl BRIEF DESCRIPTION OF THE DRAWINGS
- 0078 +p FIG. 1 is a graph reporting the residual crystallinity of <<<<>>>>exemplary elastomer blends of
- 0079 this invention as compared to a control elastomer.
- 0080 +p FIG. 2 is a graph reporting the modulus G+40 +0 of exemplary <<<<>>>>elastomer blends of this
- 0081 invention as compared to a control elastomer and two commercially <<<<>>>>available elastomers.
- 0082 +de +cl DETAILED DESCRIPTION OF THE INVENTION
- 0083 +p The ethylene/+60 -olefin blend components of this invention are <<<< >>>polymers, i.e.,
- interpolymers, of ethylene with at least one C+hd 3+l \mathcal{C} C+hd 20 +l +60 <<<>>>>-olefin (preferably an aliphatic +60 -olefin)
- 0085 comonomer, and/or a polyene comonomer, e.g., a conjugated diene, a <<<<>>>>nonconjugated diene, a
- 0086 triene, etc. The term interpolymer includes copolymers, e.g. <<<>>>>ethylene/propylene (EP), and
- 0087 terpolymers, e.g. EPDM, but it is not limited to polymers made with <<<< >>>only ethylene and one or
- two monomers. Examples of the C+hd 3+l -C+hd 20 +l +60 -olefins <<<<>>>>include propene, +b 1+l -butene, +b 4+l -methyl-+b 1+l -pentene,
- 0089 +b 1+l -hexene, +b 1+l -octene, +b 1+l -decene, +b 1+l -dodecene, +b <<<<>>>>1+l -tetradecene, +b 1+l -hexadecene, +b 1+l -octadecene
- 0090 and +b 1+l -eicosene. The +60 -olefin can also contain a cyclic <<<< >>>>structure such as cyclohexane or
- 0091 cyclopentane, resulting in an +60 -olefin such as +b 3+l -cyclohexyl-<<<>>>>+b 1+l -propene (allyl-cyclohexane) and
- 0092 vinyl-cyclohexane. Although not +60 -olefins in the classical sense <<<<

>>>of the term, for purposes of this

0093 invention certain cyclic olefins, such as norbornene and related <<<<

>>>olefins, are +60 -olefins and can be

0094 used in place of some or all of the +60 -olefins described above. <<<<

>>>Similarly, styrene and its related

0095 olefins (e.g., +60 -methylstyrene, etc.) are +60 -olefins for <<<<

>>>purposes of this invention.

- 0096 + pg, 6
- 0097 +p Polyenes are unsaturated aliphatic or alicyclic compounds <<<<>>>>containing more than four
- 0098 carbon atoms in a molecular chain and having at least two double <<<< >>>>and/or triple bonds, e.g.,
- 0099 conjugated and nonconjugated dienes and trienes. Examples of <<<<>>>>nonconjugated dienes include
- 0100 aliphatic dienes such as +b 1,4+l -pentadiene, +b 1,4+l -hexadiene, <<<<>>>>+b 1,5+l -hexadiene, +b 2+l -methyl-+b 1,5+l -hexadiene,
- 0101 +b 1,6+l -heptadiene, +b 6+l -methyl-+b 1,5+l -heptadiene, +b 1,6+l -<<<>>>>octadiene, +b 1,7+l -octadiene, +b 7+l -methyl-+b 1,6+l -octadiene,
- 0102 +b 1,13+l -tetradecadiene, +b 1,19+l -eicosadiene, and the like; <<<>>>>cyclic dienes such as +b 1,4+l -cyclohexadiene,
- 0103 bicyclo+8 +b 2.2.1+l +9 hept-+b 2,5+l -diene, +b 5+l -ethylidene-+b <<<< >>>>2+l -norbornene, +b 5+l -methylene-+b 2+l -norbornene, +b 5+l -vinyl-+b <<<< >>>>
- 0104 2+1 -norbornene.
- 0105 bicyclo+8 +b 2.2.2+l +9 oct-+b 2,5+l -diene, +b 4+l -vinylcyclohex-+b <<<< >>>>1+l -ene, bicyclo+8 +b 2.2.2+l +9 oct-+b 2,6+l -diene,
- 0106 +b 1,7,7+l -trimethylbicyclo-+8 +b 2.2.1+l +9 hept-+b 2,5+l -diene, <<<>>>>dicyclopentadiene, methyltetrahydroindene, +b 5+l -allylbicyclo+8
- 0107 +b 2.2.1+l +9 hept-+b 2+l -ene, +b 1,5+l -cyclooctadiene, and the <<<>>>>like; aromatic dienes such as +b 1,4+l -diallylbenzene,
- 0108 +b 4+l -allyl-+b 1+l H-indene; and trienes such as +b 2,3+l -<<<>>>>diisopropenylidiene-+b 5+l -norbornene, +b 2+l -ethylidene-+b
- 0109 3+l -isopropylidene-+b 5+l -norbornene, +b 2+l -propenyl-+b 2,5+l -<<<>>>>norbornadiene, +b 1,3,7+l -octatriene,
- 0110 +b 1,4,9+l -decatriene, and the like; with +b 5+l -ethylidene-+b 2+l -<<<>>>>norbornene, +b 5+l -vinyl-+b 2+l -norbornene and +b 7+l -methyl-+b

- 0111 1,6+l -octadiene preferred nonconjugated dienes.
- 0112 +p Examples of conjugated dienes include butadiene, isoprene, +b 2,<<<
- >>>3+l -dimethylbutadiene-+b 1,3,1,2+l -dimethylbutadiene-+b 1,3,1,4+l -<<<
- >>>dimethylbutadiene-+b 1,3,1+l -ethylbutadiene-+b 1,3,2+l -<<<
- >>>phenylbutadiene-+b 1,3, +l hexadiene-+b 1,3,4+l -methylpentadiene-+b 1,<<<<
- >>>3,1,3+l -pentadiene (CH+hd 3+l CH+50 CH+13 CH+50 CH+hd 2+l ; commonly
- 0113 called piperylene), +b 3+l -methyl-+b 1,3+l -pentadiene, +b 2,4+l -<<<
- >>>dimethyl-+b 1,3+l -pentadiene, +b 3+l -ethyl-+b 1,3+l -pentadiene,
- 0114 and the like; with +b 1,3+l -pentadiene a preferred conjugated diene.
- 0115 +p Examples of trienes include +b 1,3,5+l -hexatriene, +b 2+l -methyl-<<<
- >>>+b 1,3,5+l -hexatriene, +b 1,3,6+l -heptatriene,
- 0116 +b 1,3,6+l -cycloheptatriene, +b 5+l -methyl-+b 1,3,6+l -heptatriene, <<<<
- >>>+b 5+l -methyl-+b 1,4,6+l -heptatriene,
- 0117 +b 1,3,5+l -octatriene, +b 1,3,7+l -octatriene, +b 1,5,7+l -<<<
- >>>octatriene, +b 1,4,6+l -octatriene, +b 5+l -methyl-+b 1,5,7+l -<<<
- >>>>octatriene,
- 0118 +b 6+l -methyl-+b
- 0119 1,5,7+l -octatriene, +b 7+l -methyl-+b 1,5,7+l -octatriene, +b 1,4,<<<
- >>>9+l -decatriene and +b 1,5,9+l -cyclodecatriene.

- 0120 + pg, 7
- 0121 +p Exemplary copolymers include ethylene/propylene, ethylene/butene, <<<<>>>>ethylene/+b 1+l -octene,
- 0122 ethylene/+b 5+l -ethylidene-+b 2+l -norbornene, ethylene/+b 5+l -<<<>>>>vinyl-+b 2+l -norbornene, ethylene/-+b 1,7+l -octadiene,
- 0123 ethylene/+b 7+l -methyl-+b 1,6+l -octadiene and ethylene/+b 1,3,5+l -<<<>>>>hexatriene. Exemplary terpolymers include
- 0124 ethylene/propylene/+b 1+l -octene, ethylene/butene/+b 1+l -octene, <<<< >>>>ethylene/propylene/+b 5+l -ethylidene-+b 2+l -norbornene,
- 0125 ethylene/butene/+b 5+l -ethylidene-+b 2+l -norbornene, ethylene/buten<<<< >>>>e/styrene, ethylene/+b 1+l -octene/+b
- 0126 5+l -ethylidene-+b 2+l -norbornene, ethylene/propylene/+b 1,3+l -<<<< >>>pentadiene, ethylene/propylene/+b 7+l -methyl-+b
- 0127 1,6+l -octadiene, ethylene/butene/+b 7+l -methyl-+b 1,6+l -octadiene, <<<< >>>ethylene/+b 1+l -octene/+b 1,3+l -pentadiene
- 0128 and ethylene/propylene/+b 1,3,5+l -hexatriene. Exemplary tetrapolymer<<<< >>>s include
- 0129 ethylene/propylene/+b 1+l -octene/diene (e.g. ENB), ethylene/butene/+<<<>>>>b 1+l -octene/diene and
- 0130 ethylene/propylene/mixed dienes, e.g. ethylene/propylene/+b 5+l -<<< >>>ethylidene-+b 2+l -norbornene/piperylene.
- 0131 In addition, the blend components can include minor amounts, e.g. +b <<<< >>>>0.05(1-+b) 0.5 +l percent by weight,
- 0132 of long chain branch enhancers, such as +b 2,5+l -norbornadiene (aka <<<< >>>>bicyclo+8 +b 2,2,1+l +9 hepta-+b 2,5+l -diene),
- 0133 as diallylbenzene, +b 1,7+l -octadiene (H+hd 2+l C+50 CH(CH+hd 2+l <<<<>>>>)+hd 4+l CH+50 CH+hd 2+l), and +b 1,9+l -decadiene
- 0134 (H+hd 2+1 C+50 CH(CH+hd 2+1)+hd 6+1 CH+50 CH+hd 2+1).
- 0135 +p Typically, the blend components of this invention comprise at <<<<

- >>>>least about +b 20, +1 preferably
- 0136 at least about +b 30 +l and more preferably at least about +b 40, +l <<<<>>>>weight percent ethylene; at least about +b 1, +l
- 0137 preferably at least about +b 5 +l and more preferably at least about <<<<>>>>+b 10, +l weight percent of at least one +60 -olefin;
- 0138 and, if a polyene-containing terpolymer, greater than +b 0, +l <<<<>>>>preferably at least about +b 0.1 +l and
- 0139 more preferably at least about +b 0.5, +l weight percent of at least <<<< >>>one conjugated or nonconjugated
- 0140 polyene. As a general maximum, the blend components of this invention <<<< >>>>comprise not more
- 0141 than about +b 95, +l preferably not more than about +b 85 +l and more <<<< >>>preferably not more than about +b 75, +l
- 0142 weight percent ethylene; not more than about +b 80, +l preferably not <<<<>>>>more than about +b 70 +l and more

- 0143 + pg.8
- 0144 preferably not more than about +b 60, +l weight percent of at least <<<< >>>one +60 -olefin; and, if a terpolymer,
- 0145 not more than about +b 20, +l preferably not more than about +b 15 +l <<<<>>>>and more preferably not more than
- 0146 about +b 12, +l weight percent of at least one of a conjugated or <<<<>>>>nonconjugated diene. All weight
- 0147 percentages are based on weight of the blend.
- 0148 +p Important to this invention is that the difference in ethylene <<<<>>>>content between the first and
- 0149 second components of the blend is at least about +b 10 +l weight <<<<>>>>percent, preferably at least about +b 15 +l
- 0150 and more preferably at least about +b 20, +l weight percent. The <<<<>>>>maximum difference in ethylene
- 0151 content between the first and second components of the blend can vary <<<<>>>>widely although as a
- 0152 practical matter, the maximum difference does not exceed about +b 30, <<<< >>>>+l preferably about +b 25, +l weight
- 0153 percent.
- 0154 +p The ethylene/+60 -olefin polymer components of this invention can <<<< >>>be produced using
- 0155 conventional ethylene/+60 -olefin polymerization technology. <<<>>>>Preferably, the ethylene/+60 -olefin
- 0156 polymer components of this invention are made using a mono- or bis-<<<>>>>cyclopentadienyl, indenyl,
- 0157 or fluorenyl transition metal (preferably Group +b 4+l) catalysts or <<<<>>>>constrained geometry catalysts
- 0158 (CGC) in combination with an activator, in a solution, slurry, or gas <<<< >>>>phase polymerization

- 0159 process. The catalyst is preferably mono-cyclopentadienyl, mono-<<<<>>>>indenyl or mono-fluorenyl
- 0160 CGCs. The solution process is preferred. U.S. Pat. No. +b 5,064,802; <<<>>>>+1 WO+b 93/19104 +l (U.S. Ser. No. +b 8,003, +l filed
- 0161 Jan. +b 21, 1993+l), and WO+b 95/00526 +l disclose constrained <<<< >>>>geometry metal complexes and
- 0162 methods for their preparation. Variously substituted indenyl <<<<>>>>containing metal complexes are
- 0163 taught in WO+b 95/14024 +l and WO+b 98/49212. +l The relevant <<<< >>>>teachings of all of the foregoing patents
- 0164 or their corresponding U.S. patents or allowed applications are <<<< >>>>hereby incorporated by reference
- 0165 for purposes of U.S. patent practice.

- 0166 +pg,9
- 0167 +p In general, polymerization may be accomplished at conditions well <<<< >>>>known in the art for
- 0168 Ziegler-Natta or Kaminsky-Sinn type polymerization reactions, that is,<<<
>>>> temperatures from +b 0+1 -+b
- 0169 250+1 +20 +0 C., preferably +b 30+1 -+b 200+1 +20 +0 C., and <<<<>>>>pressures from atmospheric to +b 10,000 +l atmospheres (+b 1013 +l
- 0170 megapascals (MPa)). Suspension, solution, slurry, gas phase, solid <<<<>>>>state powder polymerization
- 0171 or other process conditions may be employed if desired. A support, <<<<>>>>especially silica, alumina, or
- 0172 a polymer (especially poly(tetrafluoroethylene) or a polyolefin) may <<<<>>>>be employed, and desirably
- 0173 is employed when the catalyst is used in a gas phase polymerization <<<<>>>>process. The support is
- 0174 preferably employed in an amount sufficient to provide a weight ratio <<<<>>>>of catalyst (based on
- 0175 metal):support within a range of from +b 1:100,000 +l to +b 1:10, +l <<<< >>>>more preferably from +b 1:50,000 +l to +b 1:20, +l
- 0176 and most preferably from +b 1:10,000 +l to +b 1:30. +l In most <<<< >>>polymerization reactions, the molar ratio of
- catalyst:polymerizable compounds employed is from +b 10+hu, 12+b :1 +l <<<>>>>to +b 10+hu, 1+b :1, +l more preferably from +b 10+hu +31
- 0178 9+b :1 +l to +b $10+hu_0^{43}5+b$:1.
- 0179 +p Inert liquids serve as suitable solvents for polymerization. <<<<>>>>Examples include straight and
- 0180 branched-chain hydrocarbons such as isobutane, butane, pentane, <<<<>>>>hexane, heptane, octane, and
- 0181) mixtures thereof; cyclic and alicyclic hydrocarbons such as <<<<

- >>>cyclohexane, cycloheptane,
- 0182 methylcyclohexane, methylcycloheptane, and mixtures thereof; <<<
- >>>>perfluorinated hydrocarbons
- 0183 such as perfluorinated C+hd 4-10 +l alkanes; and aromatic and alkyl-<<<>>>>substituted aromatic compounds
- 0184 such as benzene, toluene, xylene, and ethylbenzene. Suitable solvents <<<< >>>>also include liquid olefins
- 0185 that may act as monomers or comonomers including butadiene, <<<< >>>>cyclopentene, +b 1+l -hexene, +b 1+l -hexane,
- 0186 +b 4+l -vinylcyclohexene, vinylcyclohexane, +b 3+l -methyl-+b 1+l -<<<>>>>pentene, +b 4+l -methyl-+b 1+l -pentene, +b 1,4+l -hexadiene,
- 0187 +b 1+l -octene, +b 1+l -decene, styrene, divinylbenzene, allylbenzene, <<<>>>> and vinyltoluene
- 0188 (including all isomers alone or in admixture). Mixtures of the <<<<>>>> foregoing are also suitable. If

- 0189 + pg, 10
- 0190 desired, normally gaseous olefins can be converted to liquids by <<<<>>>>application of pressure and used
- 0191 herein.
- 0192 +p The ethylene/+60 -olefin polymer components of this invention can <<<< >>>>be blended by any in-reactor
- 0193 or post-reactor process. The in-reactor blending processes are <<<<>>>>preferred to the post-reactor
- 0194 blending processes, and the processes using multiple reactors <<<<>>>>connected in series are the
- 0195 preferred in-reactor blending processes. These reactors can be <<<<>>>>charged with the same catalyst
- 0196 but operated at different conditions, e.g., different reactant <<<<>>>>concentrations, temperatures,
- 0197 pressures, etc, or operated at the same conditions but charged with <<<< >>>>different catalysts.
- 0198 +p Examples of processes that can be use to form the blends of this <<<< >>>invention include the use
- 0199 of an ethylene/+60 -olefin polymerization catalyst utilized in <<<<
- 0200 additional homogeneous or heterogeneous polymerization catalyst in <<<< >>>>the same reactor or in
- 0201 separate reactors that are connected in series or in parallel to <<<< >>>>prepare polymer blends having
- 0202 desirable properties. An example of such a process is disclosed in WO <<<< >>>+b 94/00500 +l at page +b 29 +l line
- 0203 +b 4 +l to page +b 33 +l line +b 17. +l The process uses a continuous<<<<>>>>ly stirred tank reactor (CSTR) connected in
- 0204 series or parallel to at least one other CSTR or tank reactor. WO +b <<<<

>>>93/13143 +1 (at page +b 2 +1 lines +b 19+1 -+b)

0205 31+l) teaches polymerizing monomers in a first reactor using a first <<<< >>>>CGC having a first reactivity

0206 and polymerizing monomers in a second reactor using a second CGC <<<<>>>>having a second reactivity

0207 and combining the products from the two reactors. Page +b 3, +l lines <<<< >>>>+b 25+1 -+b 32 +l of WO +b 93/13143 +l

0208 provides teachings about the use of two CGCs having different <<<<>>>>reactivities in one reactor. WO

0209 +b 97/36942 +l (page +b 4 +l line +b 30 +l through page +b 6 +l line <<<<>>>>+b 7+l) teaches the use of a two-loop reactor system.

0210 The relevant teachings of such applications or their corresponding <<<<>>>>U.S. patents and allowed

0211 applications are incorporated herein by reference for purposes of <<<<>>>>U.S. patent practice.

- 0212 + pg, 11
- 0213 +p The polydispersity (molecular weight distribution or Mw/Mn or MWD) <<<<>>>> the polymer
- 0214 blend generally ranges from at least about +b 2, +l preferably at <<<<>>>>least about +b 2.1, +l and especially at least
- 0215 about +b 2.2 +l to about +b 10, +l preferably about +b 6, +l and <<<<>>>>especially about +b 4.
- 0216 +p The polydispersity index is typically measured by gel permeation <<<< >>>>chromatography (GPC)
- 0217 on a Waters +b 150+l +20 +0 C. high temperature chromatographic unit <<<< >>>equipped with three linear mixed bed
- 0218 columns (Polymer Laboratories (+b 10 +1 micron particle size)) <<<<>>>>operating at a system temperature of
- 0219 +b 140+l +20 +0 C. The solvent is +b 1,2,4+l -trichlorobenzene from <<<< >>>>which about +b 0.5+l % by weight solutions of the
- 0220 samples are prepared for injection. The flow rate is +b 1.0 +l <<<<>>>>milliliter/minute, and the injection size is
- 0221 +b 100 +l microliters.
- 0222 +p The molecular weight determination is deduced by using narrow <<<<>>>>molecular weight
- 0223 distribution polystyrene standards (from Polymer Laboratories) in <<<<>>>>conjunction with their elusion
- 0224 volumes. The equivalent polyethylene molecular weights are determined <<<< >>>>by using appropriate
- 0225 Mark-Houwink coefficients for polyethylene and polystyrene (as <<<< >>>described by Williams and
- 0226 Ward in Journal of Polymer Science, Polymer Letters, Vol. +b 6, +l <<<< >>>>(+b 621+l) +b 1968+l) to derive the equation:+ps
- 0227 +ti M+hd polyethylene+1 +32 (+i a+1)(M+hd polystyrene+1)+hu b+ps

- 0228 +ps In this equation, a+32 +b 0.4316 +l and b+32 +b 1.0. +l Weight <<<< >>>>average molecular weight, Mw, is calculated in
- 0229 the usual manner according to the formula:+ps
- 0230 +tigMw+32 +934 (w+hd i+l) (M+hd i+l)+ps
 - 0231 +ps where w+hd i +l and M+hd i +l are the weight fraction and <<<<>>>>molecular weight respectively of the ith fraction
 - 0232 eluting from the GPC column. Generally, the Mw of the polymer blend <<<< >>>>ranges from about
 - 0233 +b 10,000, +l preferably about +b 20,000, +l more preferably about +b <<<<>>>>40,000, +l and especially about +b 60,000, +l to

- 0234 + pg, 12
- 0235 about +b 1,000,000, +l preferably about +b 800,000, +l more <<<<>>>>preferably about +b 600,000, +l and especially about
- 0236 +b 500,000.
- 0237 +p The polymer blends of this invention cover a range of viscosities, <<<<>>>>depending upon the
- 0238 molecular weight of the blend and optional post-polymerization <<<<>>>>rheological modification. In
- 0239 general, the blend viscosity is characterized by a Mooney viscosity <<<<>>>>which is measured according
- 0240 to ASTM D +b 1646+l -+b 89 +l using a shear rheometer at +b 125+l +20 <<<<>>>>+0 C. The polymer blend Mooney viscosity
- 0241 generally ranges from a minimum of less than +b 0.01, +l preferably <<<<>>>>+b 0.1, +l more preferably about +b 1, +l and
- 0242 especially about +b 15 +l to a maximum of about +b 150, +l preferably <<<< >>>about +b 125, +l more preferably about
- 0243 +b 100, +l and especially about +b 70.
- 0244 +p The rheological or shear thinning behavior of the ethylene <<<< >>>>interpolymer is determined by
- 0245 measuring the ratio of interpolymer viscosity at +b 0.1 +l rad/sec to <<<< >>>viscosity at +b 100 +l rad/sec. This ratio
- 0246 is known as the Rheology Ratio (RR), V+b 0.1/+1 V+b 100, +l or more <<<>>>>simply, +b 0.1/100. +l The RR is an
- 0247 extension of I+hd 10+l /I+hd 2 +l and as such, in those instances in <<<<>>>>which the measurement of I+hd 2 +l and I+hd 10 +l are
- 0248 difficult, e.g., the I+hd 2 +l is less than +b 0.5, +l or the <<<< >>>>molecular weight of the interpolymer is relatively
- 0249 high, or the Mooney viscosity of the interpolymer is greater than <<<< >>>>about +b 35, +l the RR of the

- 0250 interpolymer can be measured using a parallel plate rheometer.
- 0251 +p The density of the polymer blends is measured according to ASTM D-<<<<>>>>+b 792, +l and this
- 0252 density ranges from a minimum of about +b 0.850 +l grams/cubic <<<<>>>>centimeter (g/cm+hu 3+l), preferably about
- 0253 +b 0.853 +l g/cm+hu 3+l , and especially about +b 0.855 +l g/cm+hu <<<<>>>>3+l , to a maximum of about +b 0.970 +l g/cm+hu 3+l , preferably
- 0254 about +b 0.940 +l g/cm+hu 3+l , and especially about +b 0.930 +l <<<<>>>>g/cm+hu 3+l . For those polymer blends that are
- 0255 elastomers, i.e., with a crystallinity less than about +b 45+l %, the <<<<>>>>maximum density is about +b 0.895, +l
- 0256 preferably about +b 0.885 +l and more preferably +b 0.875, +l g/cm+hu <<<<>>>>3+l .

- 0257 + pg, 13
- $^{0258}\,$ +p For polymer blends intended for use as elastomers, the crystallini <<<>>>> ty is preferably less than
- 0259 about +b 40, +l more preferably less than about +b 30, +l percent, <<<<>>>>preferably in combination with a melting
- 0260 point of less than about +b 115, +l preferably less than about +b 105,<<<<>>>> +l C, respectively. Elastomeric
- 0261 polymer blends with a crystallinity of zero to +b 25 +l percent are <<<<>>>>even more preferred. The percent
- 0262 crystallinity is determined by dividing the heat of fusion as <<<<>>>>determined by differential scanning
- 0263 calorimetry (DSC) a of polymer blend sample by the total heat of <<<< >>>>fusion for that polymer blend
- 0264 sample. The total heat of fusion for high-density homopolymer <<<< >>>>polyethylene (+b 100+l % crystalline)
- 0265 is +b 292 +l joule/gram (J/g).
- 0266 +p One hallmark of this invention is that a desirable property of one <<<<>>>>component of the blend
- 0267 can be enhanced without a significant diminution of one or more <<<< >>>>desirable properties of another
- 0268 component. For example, certain blends of this invention exhibit an <<<< >>>enhanced low temperature
- 0269 impact property relative to one component of the blend without any <<<< >>>significant diminution of the
- 0270 glass transition temperature (Tg) of the other component of the <<<< >>>>blend. Other blends of this
- 0271 invention exhibit the same phenomena (i.e., no significant diminution <<<< >>>>of Tg) with respect to
- 0272 pellet flow (i.e., the ability of pellets made from the blend to move <<<<

- >>>pass one another without
- 0273 sticking or blocking), compression set for a given crystallinity, <<<<>>>>melt strength and shape retention.
- 0274 +p Another hallmark of this invention is that these blends exhibit an <<<< >>>improved combination
- 0275 of low temperature, pellet flow, compression set, melt strength <<<< >>>>and/or shape retention properties
- 0276 as compared to an ethylene/+60 -olefin polymer blend of similar <<<<>>>>composition but in which the
- 0277 ethylene content of each component is substantially the same.
- $^{0278}\,$ +p The following examples are provided as a further illustration of <<<< >>>>the invention. Unless
- 0279 stated to the contrary, all parts and percentages are by weight.

- 0280 + pg, 14
- 0281 +cl Specific Embodiment
- 0282 +p Four elastomers were prepared using a dual loop reactor such as <<<< >>>>that described in WO
- 0283 +b 98/49212. +l Each elastomers was prepared under the same <<<< >>>>conditions with the same reactants and
- 0284 catalyst and to the same total ethylene content (+b 66 +l weight <<<<>>>>percent based upon the weight of the
- 0285 polymer component) as the other elastomers. The control elastomer was <<<< >>>a blend of two
- 0286 essentially identical components, i.e., the component made in the <<<< >>>>first loop reactor was
- 0287 essentially the same in composition and properties as the component <<<<>>>>made in the second loop
- 0288 reactor. The remaining three elastomers, i.e., Elastomers +b 1, 2 +l <<<<>>>>and +b 3, +l are embodiments of this
- 0289 invention. Each is essentially the same as the other and the control <<<< >>>except that the ethylene
- 0290 content of the component made in the first loop reactor is different <<<< >>>>than the ethylene content of
- 0291 the component made in the second loop reactor. The composition, <<<<>>>>Mooney viscosity, weight
- 0292 average molecular weight (Mw), molecular weight distribution (MWD), <<<<>>>>temperature of
- 0293 crystallinity (Tc, both onset and peak), and glass transition <<<<>>>>temperature (Tg) for each elastomer
- 0294 and two commercially available elastomers (Dutral+198 +0 +b 4038 +l <<<< >>>>manufactured and sold by Enichem,
- 0295 and Nordel+198 +0 IP +b 4770 +l manufactured and sold by Dupont Dow <<<<

>>>Elastomers) are reported in the 0296 following table.

- 0297 + pg, 15
- 0298 +t,0150
- 0299 +p As is evident from the data in the above table, Elastomers +b 1, 2 <<<<>>>>+1 and +b 3 +l not only have a
- 0300 lower Tg than the control elastomer, but also a lower Tg than the two <<<<>>>>commercially available
- 0301 elastomers (both of similar composition). Lower Tg usually means <<<<>>>>better low temperature
- 0302 flexibility in such products as seals, belts and automotive hoses.
- 0303 +p The residual crystallinity at elevated temperatures of Elastomers <<<<>>>>+b 1, 2 +l and +b 3 +l are compared
- 0304 with the Control Elastomer in FIG. 1. As can be seen from this graph, <<<<>>>>as the ethylene split
- 0305 between the elastomer components increases, the so does the residual <<<< >>>>crystallinity. Usually, the
- 0306 larger the residual crystallinity at higher temperatures, the better <<<< >>>>the shape retention of the
- 0307 elastomer (neat or deployed in its intended end-use).
- 0308 +p FIG. 2 reports the modulus G+40 0 of the Control Elastomer, <<<< >>>>Elastomers +b 1, 2 +l and +b 3, +l Nordel
- 0309 IP +b 4770 +l and Dutral +b 4038. +l Modulus G+b 3+l +40 , or storage <<<>>>>modulus, is another measure of the shape

- 0310 + pg, 16
- 0311 retention of the elastomer. Here too, Elastomers +b 1, 2 +l and +b 3 <<<<>>>>+l outperform the Control Elastomer
- 0312 even with a slightly higher overall ethylene content.
- 0313 +p Finally, Elastomers +b 1 +l and +b 2 +l were compared with the <<<< >>>>Control Elastomer for pellet flow.
- 0314 Elastomers +b 1 +l and +b 2 +l demonstrated superior temperature <<<<>>>>resiliency and lower blocking than the
- 0315 Control Elastomer.
- $^{0316}\,\,$ +p Although the invention has been described in considerable detail <<<< >>>>through the
- 0317 specification and examples, one skilled in the art can make many <<<<>>>>variations and modifications
- 0318 without departing from the spirit and scope of the invention as <<<< >>>>described in the following claims.

- 0319 + pg, 17
- 0320 +cm What is claimed is:
- 0321 +cm 1. An ethylene/+60 -olefin polymer blend comprising first and <<<<>>>>second ethylene/+60 -olefin
- 0322 polymer components in which the ethylene content of the first <<<<>>>>component differs by at
- 0323 least about +b 10 +l weight percent from the ethylene content of the <<<< >>>second component.
- 0324 +cm 2. The blend of claim 1 in which the ethylene content of the <<<< >>>first component differs
- 0325 by at least about +b 15 +l weight percent from the ethylene content <<<<>>>>of the second component.
- 0326 +cm 3. The blend of claim 1 in which the +60 -olefin in each <<<< >>>component contains from +b 3 +l to
- 0327 about +b 8 +l carbon atoms.
- 0328 +cm 4. The blend of claim 3 in which the +60 -olefin in each <<<< >>>>component is propylene.
- 0329 +cm 5. The blend of claim 4 in which the first and second ethylene/+6<<<<>>>>0 -olefin polymer
- 0330 components further comprise a polyene.
- 0331 +cm 6. The blend of claim 5 in which the polyene is +b 5+l -<<<>>>>ethylidene-+b 2+l -norbornene.
- 0332 +cm 7. The blend of claim 3 in which the +60 -olefin in the first <<<< >>>component is propylene
- 0333 and the +60 -olefin in the second component contains from +b 4 +l to <<<< >>>>+b 8 +l carbon atoms.
- 0334 +cm 8. The blend of claim 7 in which at least one of the first and <<<<>>>>second ethylene/+60 -olefin
- 0335 polymer components further comprises a polyene.

- 0336 +cm 9. An ethylene/(+60 -olefin polymer blend comprising first and <<<< >>>>second ethylene/+60 -olefin
- 0337 polymer components, the blend prepared by (i) contacting ethylene, an <<<<>>>>+60 -olefin, an
- 0338 activated constrained geometry catalyst and, optionally, a polyene, <<<<>>>>under polymerization
- 0339 conditions, in a first reactor to produce the first ethylene/+60 -<<< >>>olefin polymer component, (ii)
- 0340 transferring the first ethylene/+60 -olefin polymer component to a <<<<>>>>second reactor and in the
- 0341 presence of the first ethylene/+60 -olefin polymer component, (iii) <<<< >>>>contacting fresh ethylene, an +60 -olefin,

- 0342 + pg, 18
- 0343 an activated constrained geometry catalyst and, optionally, a polyene, <<<<>>>> under
- 0344 polymerization conditions to produce the second ethylene/+60 -olefin <<<<>>>>polymer component, the
- 0345 polymerizations of the first and second reactors conduct in such a <<<<>>>>manner that the ethylene
- 0346 content of the first ethylene/+60 -olefin polymer component is at <<<< >>>>least +b 10 +l weight percent different
- 0347 than the ethylene content of the second ethylene/+60 -olefin polymer <<<<>>>>component.
- $^{0348}\,$ +cm $^{10}.$ The blend of claim 9 in which the polymerization conducted in <<<< >>>>each reactor is a
- 0349 solution phase polymerization.

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